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Formation of Zr-based bulk metallic glass with large amount of yttrium addition

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ABSTRACT

In this study, we systematically studied the effects of Y addition on the glass forming ability (GFA), thermal stability and mechanical property of Zr₅₆Co₂₈Al₁₆ bulk metallic glass (BMG). The results show that two BMGs, i.e., Zr₄₉Co₂₈Al₁₆Y₇ and Zr₄₆Co₂₈Al₁₆Y₁₀ with diameters of at least 14 mm, were successfully prepared by the water quenching method. In addition, fully metallic glass ingot about 25 g can be obtained in the Zr₄₆Co₂₈Al₁₆Y₁₀ alloy by using the conventional arc-melting method. The origin of the high GFA of Zr₄₆Co₂₈Al₁₆Y₁₀ alloy was attributed to the smaller Gibbs free energy difference between the supercooled liquid state and crystalline state of the Zr₄₆Co₂₈Al₁₆Y₁₀ than that of Y-free alloy. On the other hand, the fracture strength of Zr-Co-Al-Y BMGs decreases with the increase of Y content. However, the Zr-Co-Al-Y BMGs containing 0-5 at.% Y still exhibit a high fracture strength of about 1690-1790 MPa. The fracture surface morphology of BMGs with 5-12 at.% Y shows a typical featureless mirror surface, implying the brittle deformation. This Zr-Co-Al-Y quaternary BMG could be regarded as a good candidate for developing a new series of BMGs.

1. Introduction

Bulk metallic glasses (BMG) have attracted significant attention for engineering, electronic and aerospace applications due to their unique properties, such as large elastic elongation limit, high fracture strength, excellent corrosion and wear resistance, superplasticity in supercooled liquid region, etc [1-4]. Since the first metallic glass of Au₇₅Si₂₅ was fabricated by Duwez et al. in 1960 [5], lots of BMGs with large critical size above centimeter have been successfully developed in Zr- [6], Fe-[7], Cu- [8], Pd- [9], Mg- [10], La- [11] and Ti-based [12] systems. Among these BMGs, many attentions have been paid to the Zr-based BMGs because of their excellent GFA and mechanical properties compared to other systems [6,13–16].

In the past decades, a series of studies have found that the structures and properties of BMGs are very sensitive to impurities, such as carbon, oxygen and other harmful elements, which would induce the heterogeneous nucleation and thus drastically reduce the GFA [1,2,17]. For instance, oxygen in the Zr-based BMGs alloys melts can induce the heterogeneous nucleation and growth of crystalline phases, such as Zr₂Cu, Zr₂Ni and Zr₄Ni₂O [18,19]. Similar observations have also been found in other BMG-forming alloys [14,20–22]. Therefore, high purity

of the constituent elements (> 99.9 at.% and oxygen < 250 ppm) and high vacuum (better than 6×10^{-3} Pa) are necessary for BMGs processing, especially for Zr- and Fe-based BMG-forming alloys [20,23,24]. In order to decrease the negative effect of oxygen, an efficient way is alloying by minor addition of the rare earth (RE) elements [21]. Among these RE elements, Y is one of the most useful minor addition element for enhancing the GFA and manufacturer of different BMG alloys [19,21,25-31]. Wang et al. reported that a proper Y addition (2 < Y < 4 at.%) can greatly improve the GFA of the Zr₅₅Al₁₅Ni₁₀Cu₂₀ alloy by inhibiting the precipitation of Zr₂Ni Laves phase [19]. Zhang et al. found that the critical diameter of Zr₅₀Ti₂Cu₃₈Al₁₀ BMG was increased from 5 mm to 20 mm by 2 at.% Y addition. In these studies, the Y addition is restricted to less than 6 at.% [31], but the effect of more content of Y addition on the GFA of Zrbased BMG has not been explored yet.

In the present work, a typical ternary Zr-based BMG-forming alloy (i.e. Zr₅₆Co₂₈Al₁₆) was selected as the base alloy due to its high GFA, which can be prepared up to 18 mm by tilt-pouring technique [16]. However, the critical size of Zr₅₆Co₂₈Al₁₆ BMG is just 6 mm under the injection-casting method according to our study. Herein, we reported a high content (10 at.%) of Y doped Zr-based BMG, Zr₄₆Co₂₈Al₁₆Y₁₀,

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Fig. 1. (a) The fractured surfaces of the arc-melted Y0-Y12 alloy ingots. (b) XRD patterns of the arc-melted Y10 ingot.

which can be prepared with a diameter of at least 14 mm by water quenching method and a 25 g amorphous ingot by conventional arcmelting method. Moreover, the thermal stability and mechanical property for the developed Zr-Co-Al-Y BMGs were explored. After that, the origin of the high GFA of the $Zr_{46}Co_{28}Al_{16}Y_{10}$ BMG-forming alloy was also studied from thermodynamic point of view.

2. Experimental

High-purity zirconium (98.5 wt%), cobalt (99.9 wt%), aluminum (99.9 wt%) and yttrium (99.9 wt%) were used as starting materials. Alloy ingots with nominal composition of Zr_{56-x}Co₂₈Al₁₆Y_x (at.%), where x = 0, 2, 5, 7, 10 and 12 (referred as Y0, Y2, Y5, Y7, Y10 and Y12 hereafter), were arc-melted on a water-cooled copper hearth under a Ti-gettered high purity argon atmosphere. Each ingot was about 25 g and re-melted at least five times to ensure chemical homogeneity. The weight loss of ingots by alloying was less than 0.2 wt%. For the BMG rods with diameter less than 6 mm, the master allovs were processed by injection-casting into copper mold in a high-vacuum (6 \times 10⁻³ Pa) chamber under argon atmosphere. For the Y7 and Y10 samples, alloys were produced by induction melting at 1473 K for 30 min in a quartz tube of 14 mm inner diameter with argon atmosphere and subsequent water quenching. It's to note that the 6 mm and 14 mm are the maximum sizes for injection-casting method and water quenching respectively in our laboratory.

The structure of prepared samples was identified on Rigaku X-ray diffraction (XRD) with Cu K_{α} radiation at 40 kV. Differential scanning calorimetry (DSC) (Netzsch STA 404C) was used to investigate the thermal properties of as-prepared BMGs under continuous high purity argon flow at a heating rate of 20 K/min. Aluminum pans were used in a temperature range of 550 K-830 K, but the Al₂O₃ pans were used for procedures that exceed 830 K. The detailed microstructure was examined by using high-resolution transmission election microscopy (HRTEM). The samples for TEM analysis were prepared using Precision Ion Polishing System (PIPS) with 3.0 keV Ar^+ ions under liquid nitrogen environment. Room temperature compression rods with diameter of 2 mm and height of about 4 mm size were directly cast from the melted alloy ingots. The uniaxial compression tests were carried out at an approximately strain rate of 5 \times 10 $^{-4}/s.$ The scanning electron microscopy (SEM) was used to observe the fracture surface morphology of the compressed samples.

The specific heat capacities (C_p) of the amorphous phase, supercooled liquid phase, and crystalline phase were also measured using DSC-404C by comparing with the specific heat capacity of a sapphire standard sample. All measurements were performed using argon gas at a heating rate of 5 K/min in the aluminum pans on C_p measurement mode.

3. Results



Fig. 1a shows the optical images of the cross-sectional fracture surface of the arc-melted ingots (Y0-Y12). The fracture surfaces of Y0,

Fig. 2. XRD patterns of the Y0-Y12 alloy samples with different diameters. (a) their critical diameters (D_c) for glass formation and (b) diameters larger than D_{c} .

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